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Contract N00014-86-K-0043

TECHNICAL REPORT No. 102

Electromagnetic Reaction to Molecular Relaxation and Its Effect on Absorption Near a Rough Surface

by

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Prepared for Publication

in

Solid State Communications

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State University of New York at Buffalo
Buffalo, New York 14260

June 1989

UNCLASSIFIED
SECURITY CLASSIFICATION OF THIS PAGE

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1a. REPORT SECURITY CLASSIFICATION Unclassified	1b. RESTRICTIVE MARKINGS
2a. SECURITY CLASSIFICATION AUTHORITY	3. DISTRIBUTION/AVAILABILITY OF REPORT Approved for public release; distribution unlimited
2b. DECLASSIFICATION/DOWNGRADING SCHEDULE	
4. PERFORMING ORGANIZATION REPORT NUMBER(S) UBUFFALO/DC/89/TR-102	5. MONITORING ORGANIZATION REPORT NUMBER(S)
6a. NAME OF PERFORMING ORGANIZATION Depts. Chemistry & Physics State University of New York 6b. OFFICE SYMBOL (If applicable)	7a. NAME OF MONITORING ORGANIZATION
6c. ADDRESS (City, State, and ZIP Code) Fronczak Hall, Amherst Campus Buffalo, New York 14260	7b. ADDRESS (City, State, and ZIP Code) Chemistry Program 800 N. Quincy Street Arlington, Virginia 22217
8a. NAME OF FUNDING/SPONSORING ORGANIZATION Office of Naval Research 8b. OFFICE SYMBOL (If applicable)	9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER Contract N00014-86-K-0043
8c. ADDRESS (City, State, and ZIP Code) Chemistry Program 800 N. Quincy Street Arlington, Virginia 22217	10. SOURCE OF FUNDING NUMBERS PROGRAM ELEMENT NO. / PROJECT NO. / TASK NO. / WORK UNIT ACCESSION NO.

11. TITLE (Include Security Classification)
Electromagnetic Reaction to Molecular Relaxation and Its Effect on Absorption Near a Rough Surface

12. PERSONAL AUTHOR(S)
Purna C. Das, Ashok Puri and Thomas F. George

13a. TYPE OF REPORT	13b. TIME COVERED FROM ____ TO ____	14. DATE OF REPORT (Year, Month, Day)	15. PAGE COUNT
		June 1989	14

16. SUPPLEMENTARY NOTATION
Prepared for publication in Solid State Communications

17. COSATI CODES			18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number)	
FIELD	GROUP	SUB-GROUP	ELECTROMAGNETIC REACTION	NEAR ROUGH SURFACE
			MOLECULAR RELAXATION	SURFACE PLASMONS
			ABSORPTION (AU)	DELOCALIZED

19. ABSTRACT (Continue on reverse if necessary and identify by block number)

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20. DISTRIBUTION/AVAILABILITY OF ABSTRACT	21. ABSTRACT SECURITY CLASSIFICATION
☒ UNCLASSIFIED/UNLIMITED ☒ SAME AS RPT ☐ DTIC USERS	Unclassified
22a. NAME OF RESPONSIBLE INDIVIDUAL Dr. David L. Nelson	22b. TELEPHONE (Include Area Code) (202) 696-4410 22c. OFFICE SYMBOL

DD Form 1473, JUN 86 *Previous editions are obsolete.*

SECURITY CLASSIFICATION OF THIS PAGE
UNCLASSIFIED

Solid State Communications, in press

ELECTROMAGNETIC REACTION TO MOLECULAR RELAXATION AND ITS EFFECT ON ABSORPTION NEAR A ROUGH SURFACE

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Abstract

The effect of electromagnetic reaction to relaxation of a molecule adsorbed on a rough surface is considered. We include the reaction fields due to the system dipole relaxing by both photon radiation and decay to delocalized surface plasmons in the determination of absorption by the adsorbate. Surface plasmon damping is shown to be important for both small- and large-sized roughness features.

PACS Nos.: 42.50kb, 68.35.Bs, 82.65.Pa

1. Introduction

Following the discovery of surface-enhanced Raman scattering (SERS),[1,2] there has been a surge in studies of surface-induced linear and nonlinear optical phenomena. Among them, surface-induced photochemistry[3-6] has received a great deal of attention. In a photochemical process, the primary step is absorption by molecules from an incident laser field and consequent excitation to a higher electronic state, followed by either dissociation or relaxation to a lower state, which may include both radiative and nonradiative mechanisms. The relative rate of dissociation determines whether the molecules undergo any significant chemical decomposition. As in SERS, the presence of a nearby surface modifies the local electric field and thus affects both the primary absorption and the relaxation steps.[4-10] The increase in the local electric field is understood to be due to resonant excitation of radiative plasma oscillations of the surface.[1,2,7,8] There may be additional competing, but mitigating, line-broadening effects due to energy transfer to the nearby surface from the excited molecules.[1-4,10,11]

Electromagnetic calculations, in the context of SERS, for a single molecule near a small metallic particle or a rough surface show that the transfer of energy between the molecule and the surface takes place via excitation of electronic modes of the associated particulate structure.[7,8] Radiative decay of these electronic excitations restores partially the energy lost by the molecule. However, in the case of a rough surface, the remaining energy is gone permanently by resistive losses in the particle and by the resonant coupling of the electronic modes to the delocalized surface plasmon modes of the underlying substrate.[9] In the case of a single ellipsoidal bump protruding out of a semi-infinite flat plane[9] as the model for a rough surface, the latter coupling mechanism and corresponding energy loss have relatively

large rates (an order or two larger in magnitude compared to the radiative and resistive decay rates) for large-sized bumps.

In view of this, we have studied the relative effects of including the reaction fields due to the decay of the molecule-bump system to surface plasmons as well as radiation damping in the electromagnetic calculation of absorption near a rough metal surface. The importance of the reaction field due to photon radiation by a system consisting of a single molecule near an ellipsoidal particle was first recognized by Wokaun et al[12] in the context of SERS. The primary effect of this radiation damping is a decrease in the local-field enhancement and hence a decrease in the absorption enhancement for large-sized particles. Because the decay to delocalized surface plasmons is the dominant mechanism for large-sized bumps,[9] a molecule in the vicinity of the bump would experience the effect of this decay channel via the reaction field. It is the intent of this paper to study the effects of these two reaction fields, corresponding to photon radidation and decay to surface plasmons on absorption enhancement.

2. Model System

A popular model for a rough surface is a hemispheroidal bump protruding out of a semi-infinite flat plane. The system, consisting of a single polarizable molecule located at a distance H from the bump surface and irradiated by an incident laser field $\vec{E}_0$, has been treated in the literature.[7,9,10] In spheroidal coordinates (ξ, η, ϕ), the hemispheroid with aspect ratio a/b is characterized[7] by a shape parameter $\xi_0 = a/f$, where $f = (a^2 - b^2)^{1/2}$. The material characteristics of the substrate are accounted for through its dielectric function $\epsilon(\omega)$.

The incident laser field $\vec{E}_0$ is taken along the z-axis, $\vec{E}_0 = E_0\hat{z}$, so that the induced dipole $\vec{\mu}$ and local field $\vec{E}_{loc}$ are aligned along the $\hat{z}$-direction. E_{loc} consists of several parts and is given by[7,9]

$$E_{loc} = E_0 - (1/f) \sum_n B_n Q_n'(\xi_1) \quad , \tag{1}$$

where $\xi_1 = (a+H)/f$, Q_n are the Legendre functions of the second kind, and the "prime" indicates the first derivative with respect to the argument ξ_1. The incident field $E_0(\omega)$ induces a dipole moment in the molecule and also polarizes the bump-plane system, which in turn produces an electric field E_R at the location of the dipole. The near field of the dipole polarizes the bump-plane system, creating an "image" dipole which generates a field E_I at the molecular dipole location.[1,7] Both E_R and E_I are contained in the second term on the right-hand side of Eq. (1).

The molecule-bump system may be imagined as a single dipole, of moment D, located near a flat surface, where $D = \mu + B_1 f^2/3$. The excited system dipole D then relaxes radiatively by emitting photons and nonradiatively by coupling to the delocalized surface plasmons of the flat substrate. The power dissipation into either of the above decay channels have been calculated by Das and Gersten.[9]

We account for the energy loss of the system by adding the radiation reaction fields E_{rad} (for photon emission) and E_{sp} (for coupling to surface plasmons) to the local electric field. To maintain self consistency we write

$$E_{loc} = E_0' - \underline{B}\cdot\underline{q} \tag{2}$$

and

$$\underline{B} = \underline{B}(\mu, E_0') \quad , \tag{3}$$

where $E_0' = E_0 + E_{rad} + E_{sp}$, $q_n = (1/f)Q_n'(\xi_1)$, and $\underline{B}$ represents a formal solution for the coefficients B_n in vector form.

The reaction fields associated with both photon radiation and delocalized surface-plasmon excitation by the system dipole, $D = D(\mu, E_0')$, may be obtained from the power radiated to photons (P_{rad}) and power lost to the excitation of surface plasmons (P_{sp}) by the system dipole. From Ref. 9,

$$P_{rad} = \frac{1}{3}|D|^2 \frac{\omega^4}{c^3}\left[1 + \frac{3}{2}\int_0^{\pi/2} \frac{d\theta \sin^3\theta[(\epsilon^2+1)\cos^2\theta + (\epsilon+1)]}{(\epsilon-1)[(\epsilon+1)\cos^2\theta - 1]}\right] \quad , \tag{4}$$

and

$$P_{sp} = 4\pi\omega |D|^2 \left(\frac{\omega}{c}\right)^3 \frac{(-\epsilon)^3}{(-\epsilon-1)^{5/2}(1-\epsilon)} \quad , \tag{5}$$

where ω, the frequency of incident photons, is taken to be resonant with the surface-plasmon excitation frequency. The fields E_{rad} and E_{sp} are determined by using the expressions for power loss given in Eqs. (4) and (5) and demanding that the work done by the radiation reaction force on the molecular dipole is equal to the negative of the energy radiated during a given time.[12] This calculation yields

$$E_{rad} = \frac{iD}{3}\left(\frac{\omega}{c}\right)^3 \left(1 + \frac{3}{2}\int_0^{\pi/2} d\theta \frac{\sin^3\theta[(\epsilon^2+1)\cos^2\theta + (\epsilon-1)]}{(\epsilon-1)[(\epsilon+1)\cos^2\theta - 1]}\right) \tag{6}$$

and

$$E_{sp} = 4\pi i D \left(\frac{\omega}{c}\right)^3 \frac{(-\epsilon)^3}{(-\epsilon-1)^{5/2}(1-\epsilon)} \quad . \tag{7}$$

For a Drude-type molecule driven by the local field E_{loc}, the power absorbed is given by

$$P = -\frac{\omega}{2} \mathrm{Im}\,(\mu^* K_1 E_0) \quad , \tag{8}$$

where

$$\mu = \frac{\omega_0^2 \alpha_0 K_1 E_0}{(\omega_0^2 - \omega^2 - i\omega\gamma)} \tag{9}$$

is a self-consistent expression for the induced dipole moment of the molecule. Part of the local field goes to shift and broaden the excited level of the molecule and appears through the modified width $\gamma = \gamma_0 + \gamma_s$. In Eq. (9), ω_0, α_0 and γ_0 are the natural frequency, static polarizability and natural line width of the molecule in the absence of the surface, respectively. The power P_0 absorbed in the absence of the surface is obtained by taking $K_1 = 1$ and $\gamma = \gamma_0$ in Eqs. (8) and (9). The ratio $R_A = P/P_0$ will be called the enhancement factor for absorption near a rough surface.

3. Results

Figure 1 shows the absorption enhancement ratio R_A for a molecule located 40 a.u. from the surface of a silver hemispheroidal bump on a flat silver

surface (the model rough surface) as a function of the semimajor axis a of the hemispheroid for various aspect ratios a/b. Optical constants for silver were obtained from Ref. 13. Numerical calculations are done for the molecule resonating with both the incident laser frequency ω as well as with the ground-state resonance frequency of the bump. For a/b = 2,3 and 4, the ground states correspond to $\mathrm{Re}\epsilon(\omega) = -7.34, -11.3$ and -15.9, respectively.[9] The frequencies associated with the ground states are obtained by fitting the optical data of Ref. 13 to a free-electron-like expression $\mathrm{Re}\epsilon(\omega) = A + B/\omega^2$. Other molecular parameters used in the numerical analysis are $\gamma_0 = 10^{-3}$ a.u. and $\alpha_0 = 10$ a.u. These graphs show the effects of both including and excluding the reaction fields in the calculations. Let us denote the contributions to the reaction fields due to photon radiation and decay to surface plasmons by the system dipole D by u_{rad} and u_{sp}, respectively. The dashed curves are obtained for the case where the reaction contributions are neglected, i.e., $u = u_{rad} + u_{sp} = 0$, and the solid curves are for $u \neq 0$. For a given bump, (a/b fixed) the absorption enhancement with $u \neq 0$ is substantial for small values of a, but rapidly decreases to values below those for $u = 0$ with increasing a, with a critical a at which both become equal. This is understandable since the effects of including the reaction fields ($u \neq 0$) are significant for large-sized roughness features (a large with a/b fixed).[12]

As mentioned earlier, u consists of two parts: u_{rad} corresponds to the inclusion of photon radiation reaction and u_{sp} to the inclusion of reaction of the decay of the system dipole to delocalized surface plasmons, which is the primary relaxation mechanism very close to the surface. The effect of only $u_{rad} \neq 0$ was discussed in Ref. 12 in the context of SERS. Curve c in Fig. 2 represents the effect of including only u_{rad} in the determination of absorption enhancement. This is in qualitative agreement with the results of Ref. 12.

For smaller-sized bumps, the effect of adding radiation damping is not significant, whereas for larger-sized bumps the enhancement is limited by the addition of the radiation damping field in the local field. Since we are not comparing our results to any realistic experimental observation, we cannot draw a quantitative conclusion except to say that the qualitative trends are in agreement with previous results in the literature.[12] On the other hand, for optical absorption by a molecule near a rough silver surface, the effect of including the surface plasmon reaction field is substantial for both intermediate and larger-sized bumps. This is shown in Fig. 2 (curves a and c). It clearly illustrates the importance of taking into consideration the reaction field corresponding to excitation of surface plasmons, which is the main thrust of this paper. It is not our intention to project curve a in Fig. 2 to be monotonically increasing with decreasing a. A portion of it (for values of $a < 300$ a.u.) is simply not shown for the relative scales involved in order to provide a qualitative comparison among the three curves. In fact, the full nature of curve a (Fig. 2) is depicted in Fig. 3 (curve d), which shows the quenching effect of $u_{sp} \neq 0$.

In Fig. 3, we plot R_A as a function of a for various fractions of u_{sp} included in u. We write $u = u_{rad} + xu_{sp}$ where $x = 0.2$, 0.3, 0.5 and 1.0 corresonding to curves a, b, c and d, respectively. Aside from the quenching in absorption observed with increasing x, there is a movement of the peak position to smaller a-values. This gives a better understanding of the enhancement of optical absorption near a rough surface. If one wants to produce a predetermined absorption enhancement (consequently a similar photodissociation enhancement) in a molecule held at a fixed distance from the surface, the surface morphology has to be designed accordingly. In other

words, the surface preparation is of paramount importance. On the other hand, if the surface roughness is specified, one needs to put the molecule at a certain distance away to obtain the maximum enhancement. For accurate prediction of spectroscopic properties of adsorbates, one has to account for the reaction field due to surface plasmon excitation within the theory. Given the surface preparation techniques known to date, it is perhaps possible to test the predictions made here regarding inclusion of reaction fields due to surface-plasmon excitation in the calculations.

4. Conclusion

In conclusion, we have considered a correction to the electromagnetic theory of surface-enhanced absorption by taking into account the reaction fields due to both radiation and decay to delocalized surface plasmon. We chose a model where decay to delocalized plasmons of the flat surface, besides photon emission, by the system is possible. It is found that for small-sized roughness features, the radiation damping correction is unimportant, but the surface plasmon reaction is important for both small- and large-sized features. It should be pointed out that the rough surface considered is only a model surface. Realistic rough surfaces are quite different and difficult to model. Predictions of this calculation may be checked experimentally on microlithographically-prepared surfaces.[14]

Acknowledgments

This research was supported in part by the National Science Foundation under Grant CHE-8620274, the Office of Naval Research and the Air Force Office of Scientific Research (AFSC), United States Air Force, under Contract F49620-86-C-0009. The United States Government is authorized to reproduce and distribute reprints for governmental purposes notwithstanding any copyright notation hereon.

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Figure Captions

Figure 1. R_A as a function of the semi-major axis a. The solid curves are for $u \neq 0$ and the dashed curves for $u = 0$. The resonance frequencies for the ground-state resonances of the silver bumps of aspect ratios $a/b = 2$, 3 and 4 are 0.1, 0.086 and 0.076 a.u., respectively. The molecule-spheroid distance is $H = 40$ a.u.

Figure 2. R_A as a function of a for fixed $a/b = 3$. Curves a and b correspond to $u \neq 0$ and $u = 0$, respectively. Curve c is obtained when only the photon radiation reaction field is taken into account ($u_{rad} \neq 0$, $u_{sp} = 0$). The molecule-spheroid separation is $H = 40$ a.u., $\epsilon(\omega_0) = -(11.3) + i(0.24)$ and $\omega_0 = 0.086$ a.u.

Figure 3. R_A as a function of a for fixed $a/b = 3$. Curves a-d correspond to $x = 0.2$, 0.3, 0.5 and 1.0, respectively, where $u = u_{rad} + xu_{sp}$. The other parameters are the same as in Fig. 2.

Fig. 1

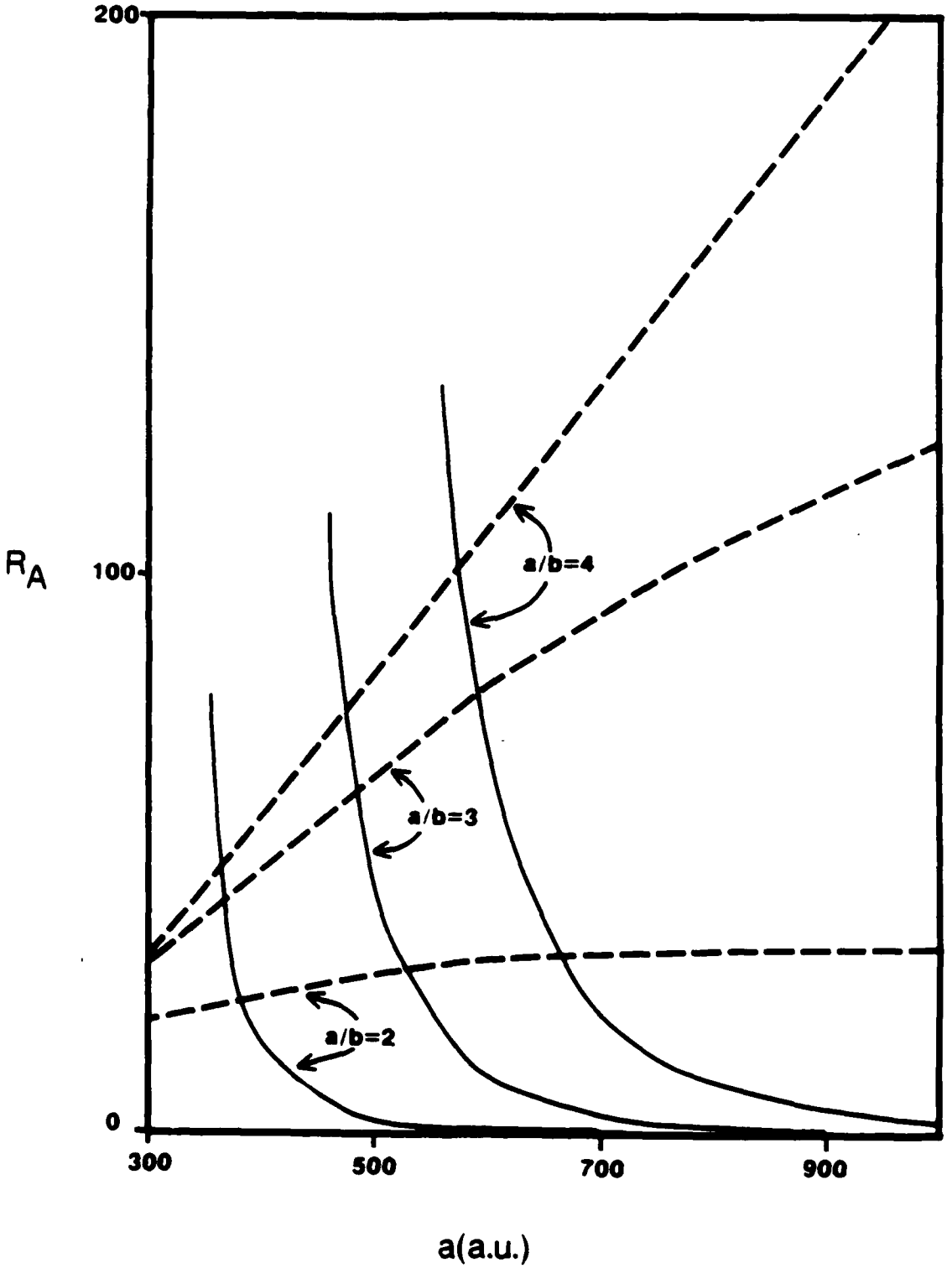


Fig.

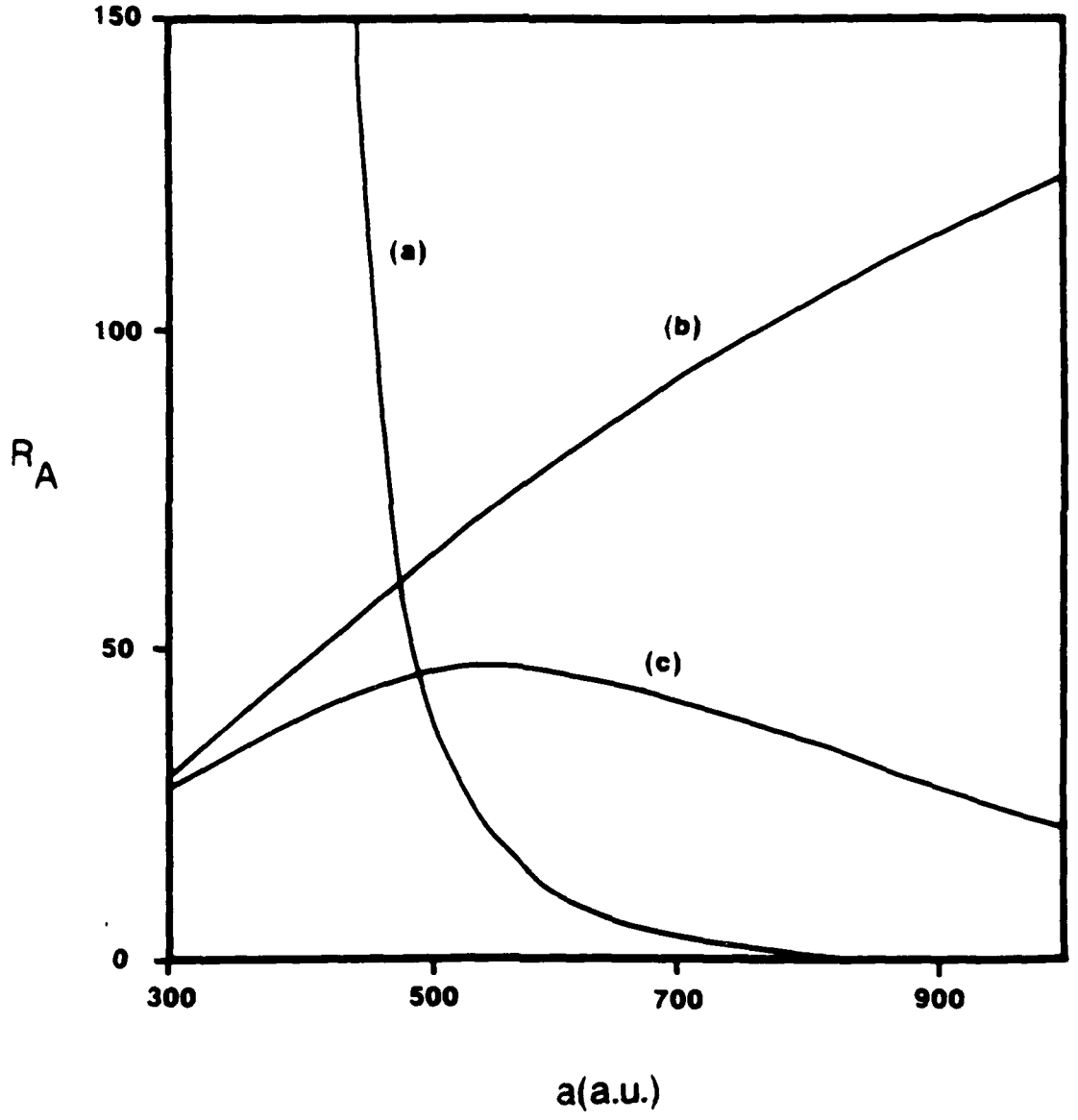
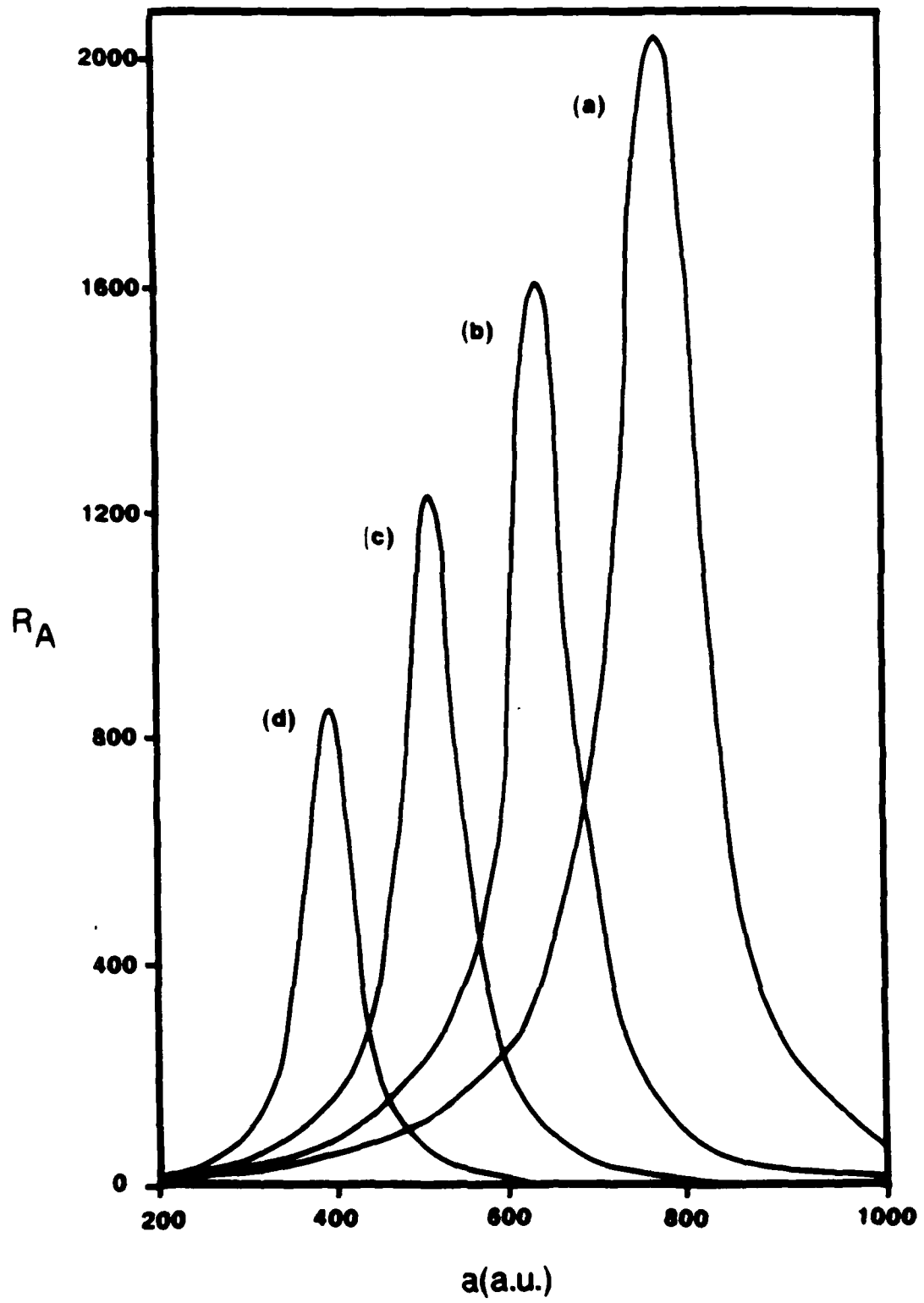


Fig. 3



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